U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Advanced Sensors and Instrumentation

Irradiation of Optical Components of *In-Situ* Laser Spectroscopic Sensors

Advanced Sensors and Instrumentation (ASI) Annual Program Webinar

October 24 – 27, 2022

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University of Michigan

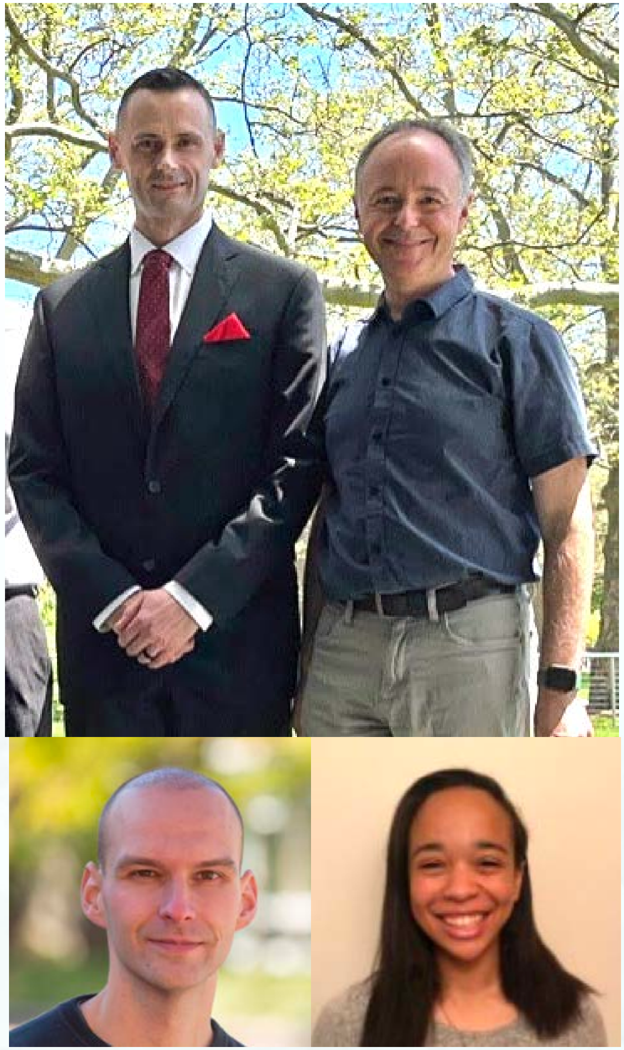
Project Overview

Goal and Objective: understand the effect of radiation damage on the performance of optical spectroscopic sensors with special emphasis on:
(1) nonlinear refractive index
(2) transient radiation-induced absorption
(3) concurrent radiation damage and thermal annealing

Schedule:

- Year 1: Procure samples; develop mobile PIE system
- Year 2: Evaluate neutron activation; construct and test heating setup; conduct gamma irradiation with post-heating
- Year 3: Conduct neutron irradiation with post-heating
- Year 4: Conduct gamma and neutron irradiation with concurrent heating

Research Team and Collaborations



Igor Jovanovic, Bryan Morgan, Londrea Garrett, Milos Burger (UM)

Piyush Sabharwall (INL)

Paul Marotta (MicroNuclear)

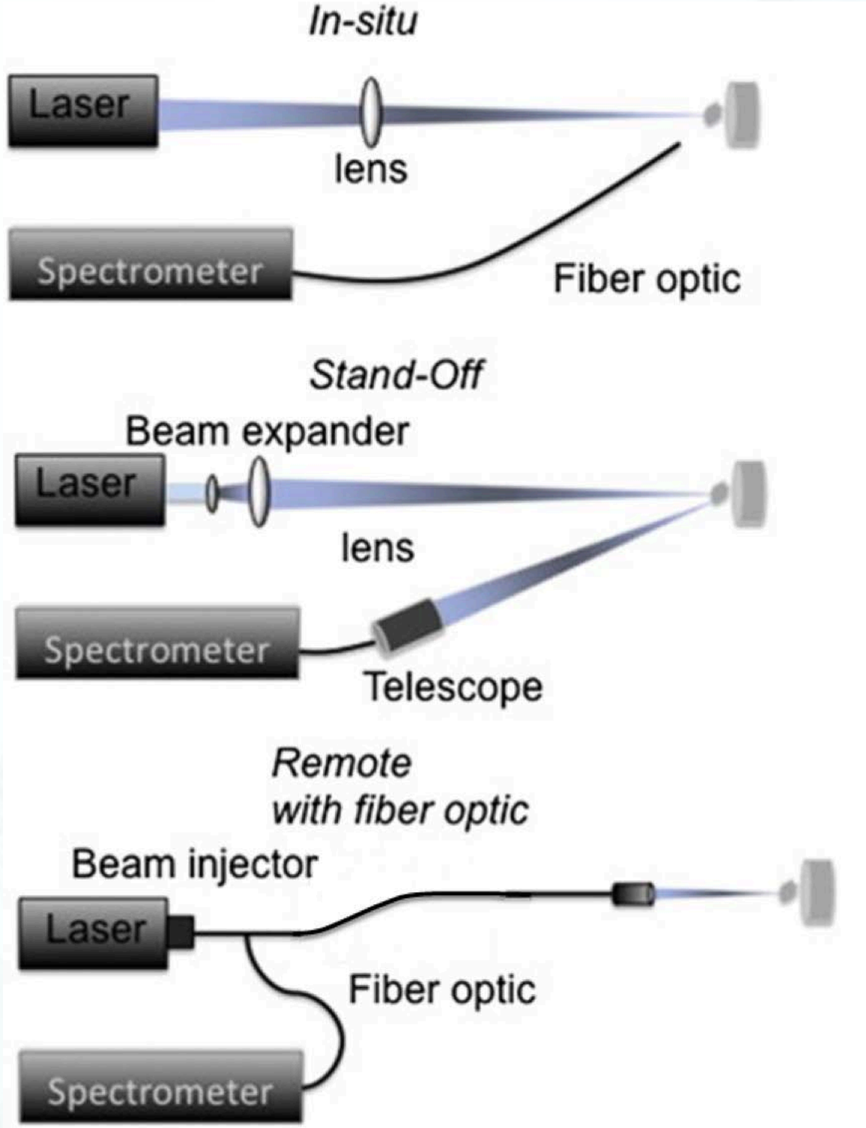
Lei Cao (OSU-NRL: NSUF)

Sungyeol Choi (Seoul National University – INERI collaborator)

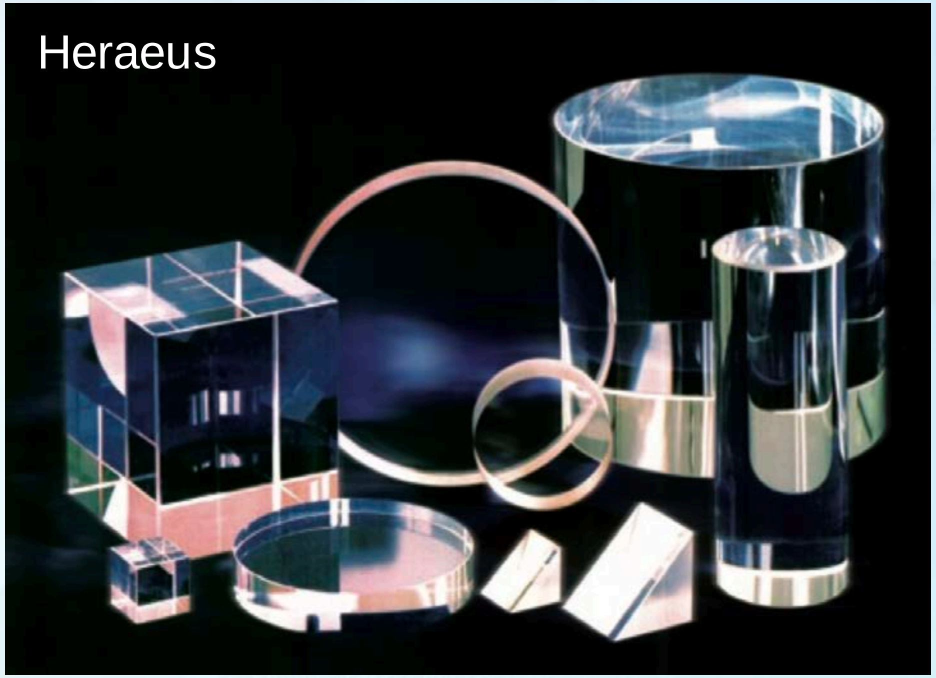
Christian Petrie (ORNL – collaborator)

Sylvain Gerard (Université Jean Morret – Saint-Étienne - collaborator)

Materials for Optical Sensors



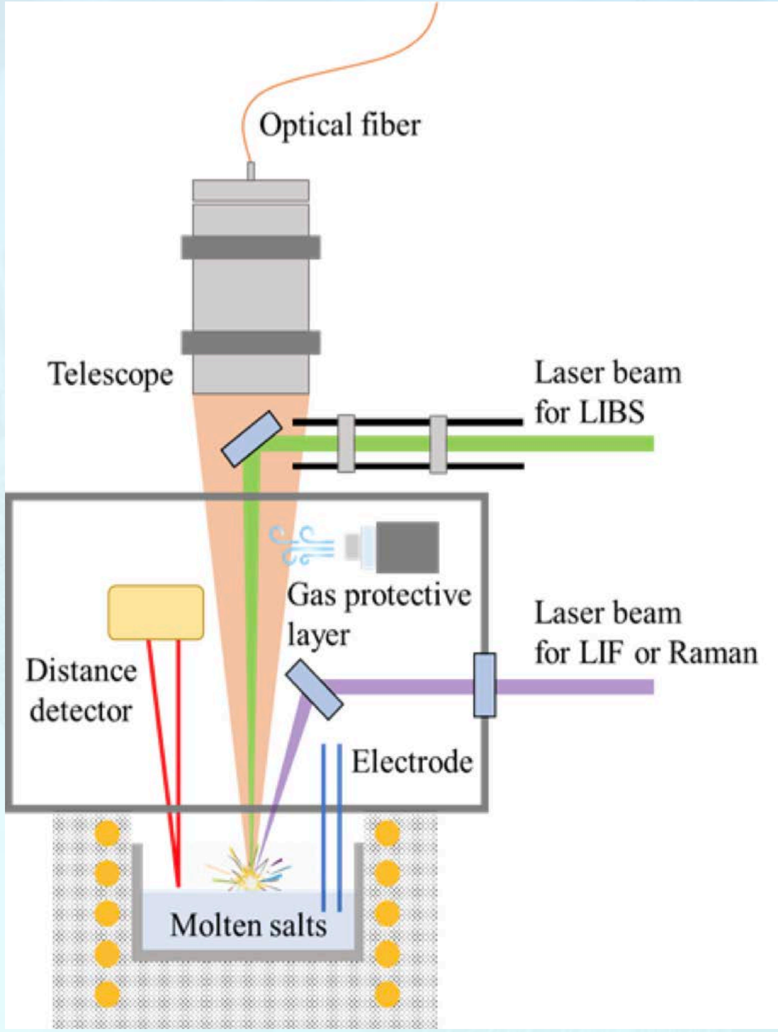
Senesi, G. S., Earth-Science Reviews 139 (2014)



Optical windows and fibers interface the instrument with the reactor coolant, transmit high intensity laser pulses, and transmit laser induced breakdown plasma spectra.

Technology Impact

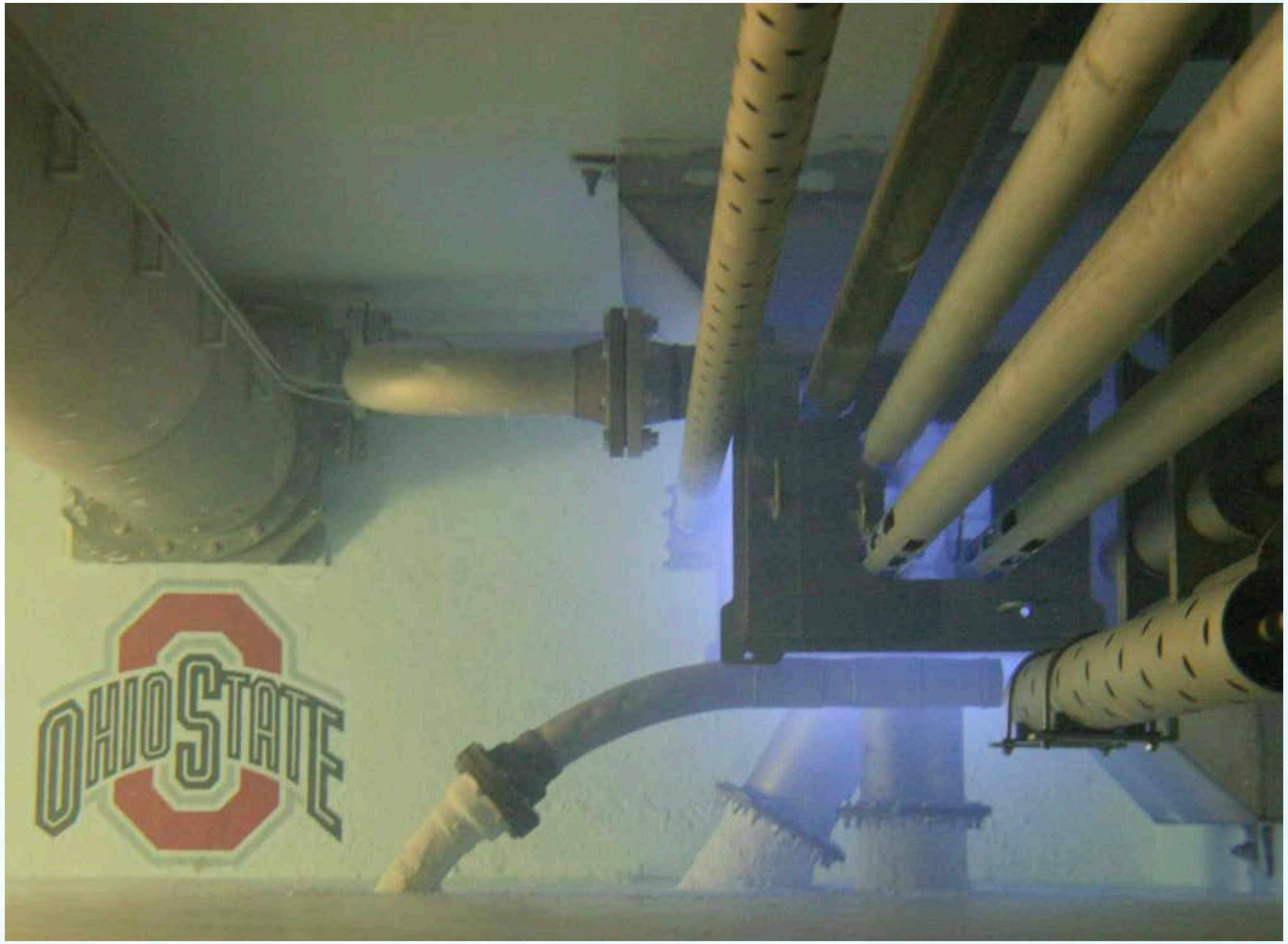
- Develop an improved understanding of radiation damage in optical materials in conditions relevant for their operation in real-time optical sensors
- First-ever attempt to quantify the effect of irradiation on nonlinear optical properties of materials
- Real-time, *in-situ* measurements of important operational parameters in advanced nuclear systems → **safety + economic performance**
- **Cross-cutting impact: design and concept of operation for a wide range of optical instrumentation in nuclear applications**



LIBS of molten salts

Irradiation Facilities

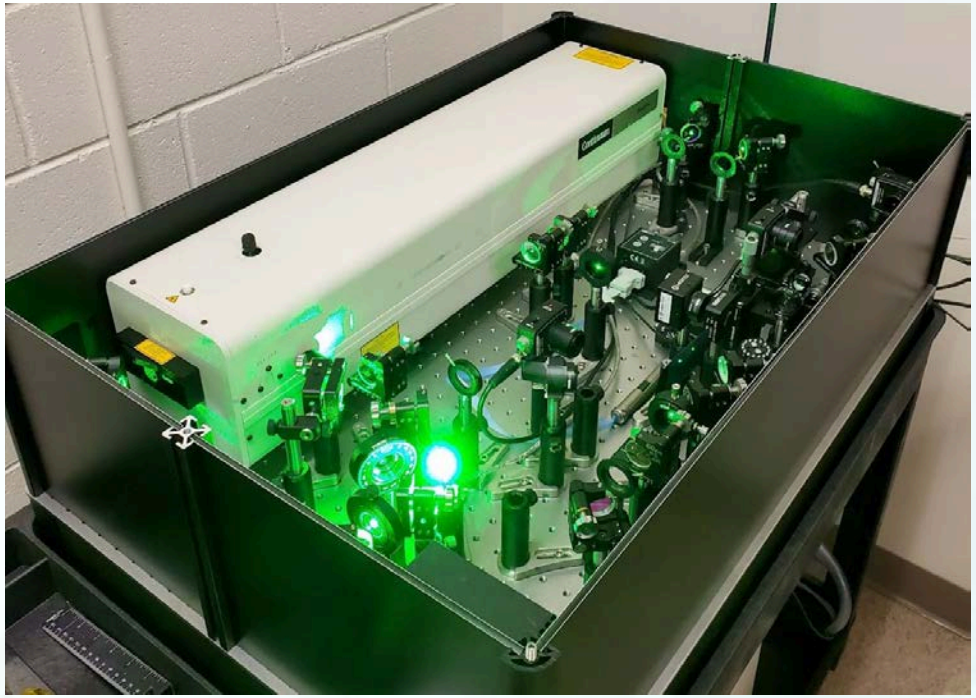
- Sample irradiation and thermal annealing at the OSU Nuclear Reactor Laboratory (DOE NSUF)
- Gamma irradiation at the PSU Radiation Science & Engineering Center (DTRA IIRM-URA)



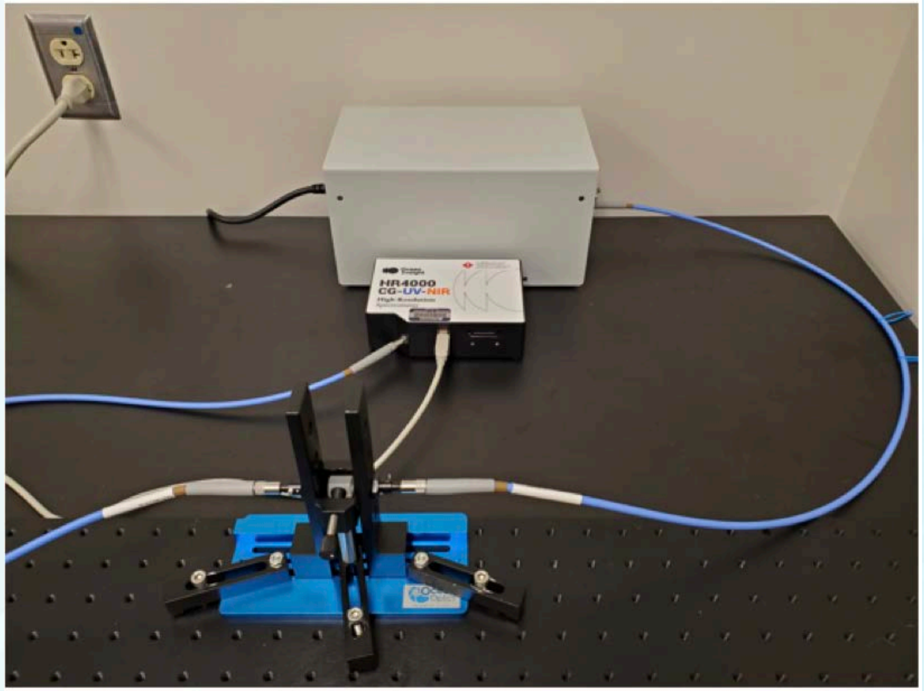
OHIO STATE

PennState

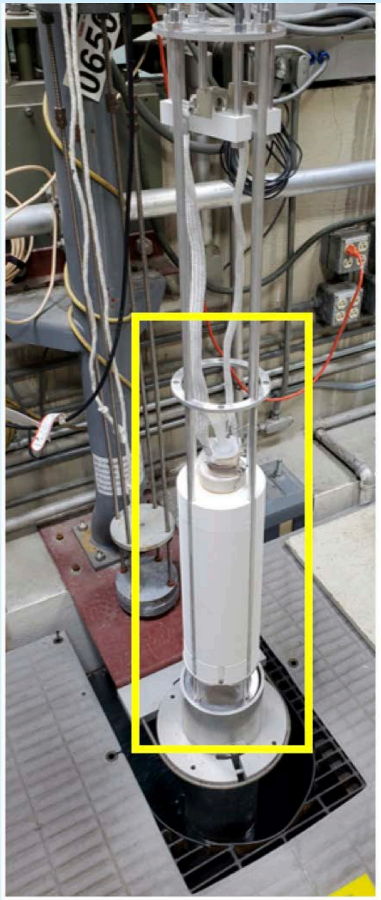
Experimental Setup



PIE: Z-scan



PIE: absorption

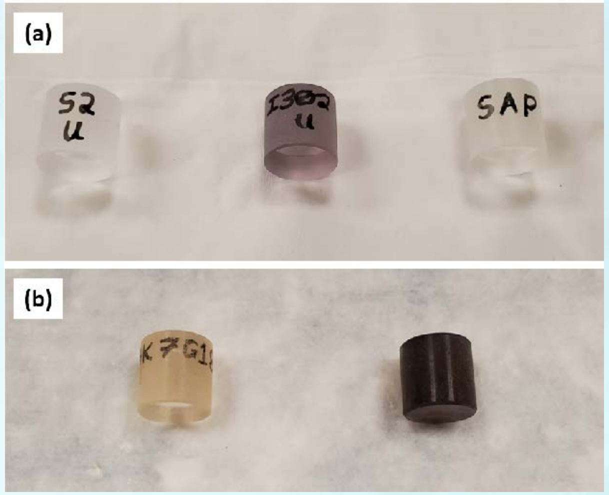


Sample furnace

Irradiation Conditions and Samples

Source	Dose/Fluence	Anneal Type	Temp.	Time
^{60}Co	600 krad 1.2 Mrad 3.4 Mrad	Post	200 °C 400 °C 600 °C 800 °C	30 min. 30 min. 30 min. 30 min.
^{60}Co	600 krad 1.2 Mrad 3.4 Mrad	Concurrent	800 °C	Duration
Reactor	3.4×10^{16} n · cm^{-2} (42 Mrad) 1.7×10^{17} n · cm^{-2} (211 Mrad)	Post	200 °C 400 °C 600 °C 800 °C	30 min. 30 min. 30 min. 30 min.
Reactor	3.4×10^{16} n · cm^{-2} (42 Mrad) 1.7×10^{17} n · cm^{-2} (211 Mrad)	Concurrent	800 °C	Duration

Material	Vendor	Type	OH Content
High-OH Fused Silica	Heraeus	Spectrosil 2000	$\leq$ 1300 ppm
Low-OH Fused Silica	Heraeus	Infrasil 302	$\leq$ 8 ppm
Sapphire	Guild Optical Associates	Optical Grade	



Recent Irradiations and Remaining PIE

- **All irradiations at OSU-NRL complete**

- Perform Z-scan and linear absorption measurements and analysis of optical windows for final irradiation fluence of ~$5x10^{17}$ n/cm^2 (633 Mrad γ)

Collaboration with Prof. Sylvain Girard – final irradiations at OSU:

•#1 iXblue F-doped SMF (acrylate coating) - radiation hardened optical fiber
•#2 iXblue F-doped SMF (polyimide coating) - radiation hardened optical fiber, for higher temperature
•#3 iXblue F-doped SMF (acrylate coating + carbon coating) ==> designed to prevent hydrogen diffusion in harsh environments combining radiation and hydrogen constraints (nuclear waste)
•#4 iXblue HACC-Ge, germanosilicate SMF (carbon coating)

UNIVERSITÉ JEAN MONNET SAINT-ÉTIENNE
LABORATOIRE HUBERT CURIEN

Nonlinear Refractive Index and Absorption

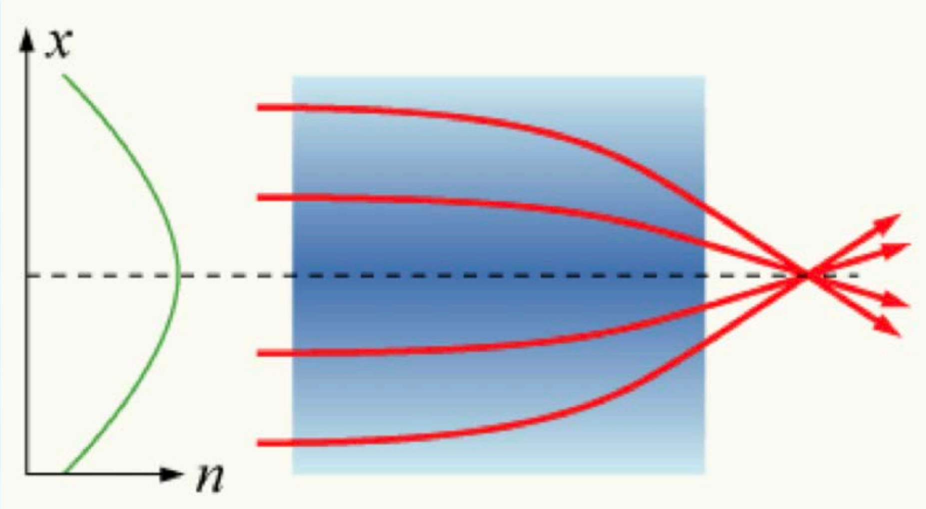
Materials exhibit nonlinear optical properties caused by variation of induced electronic polarization (P) with the applied electric field (E)

$$P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \boxed{\epsilon_0 \chi^{(3)} E^3} + \dots$$

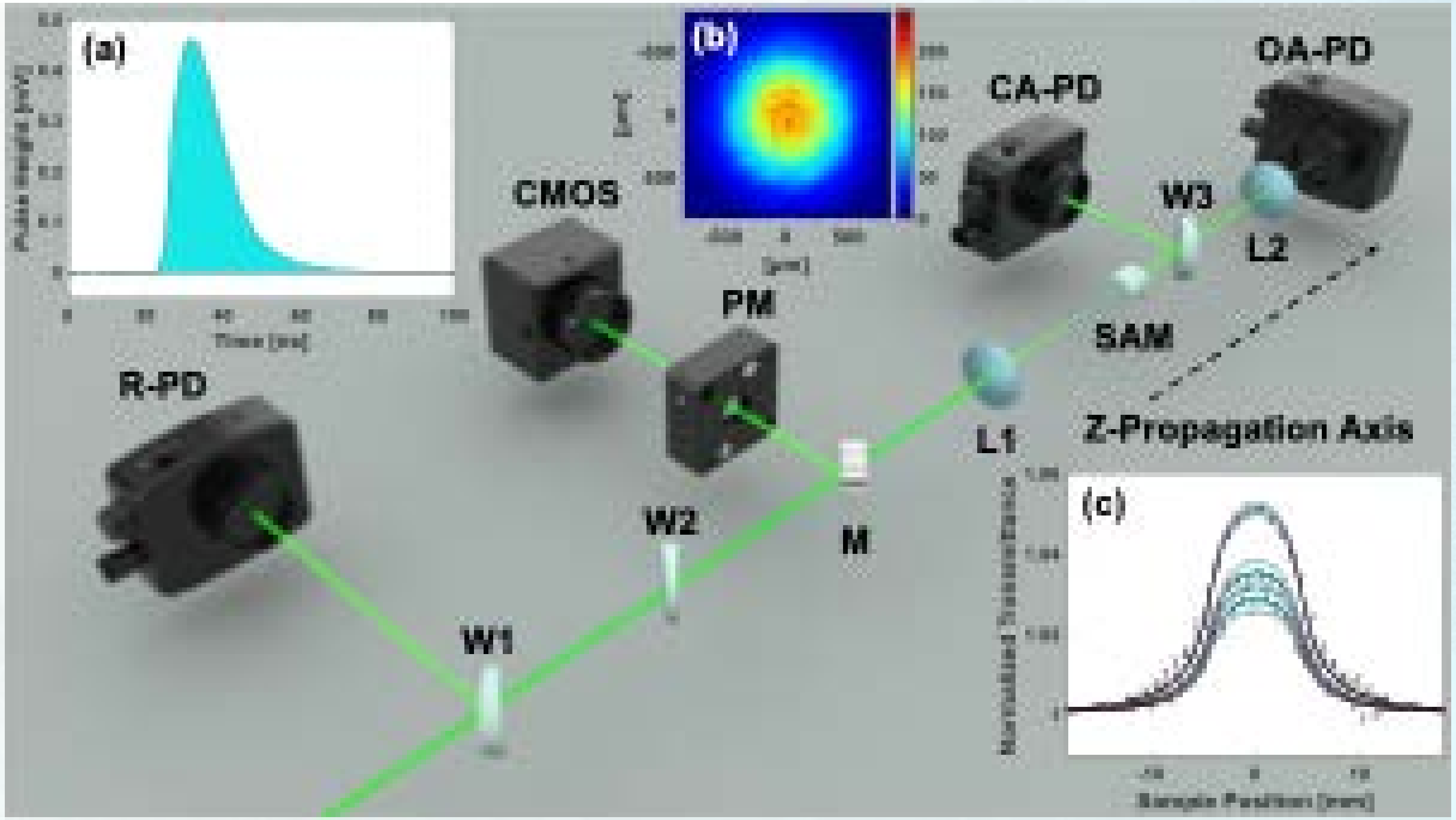
The third-order nonlinear susceptibility leads to processes such as third-harmonic generation, two-photon absorption, and the intensity-dependent refractive index.

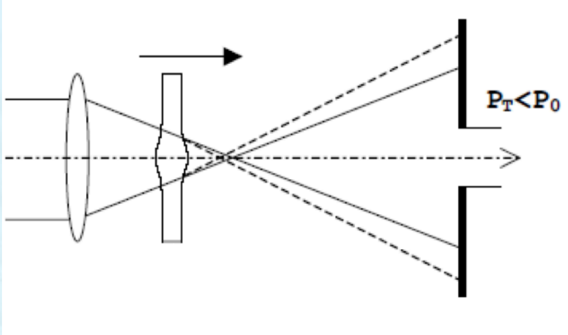
$$n = n_0 + n_2 I$$ { self-focusing / nonlinear absorption

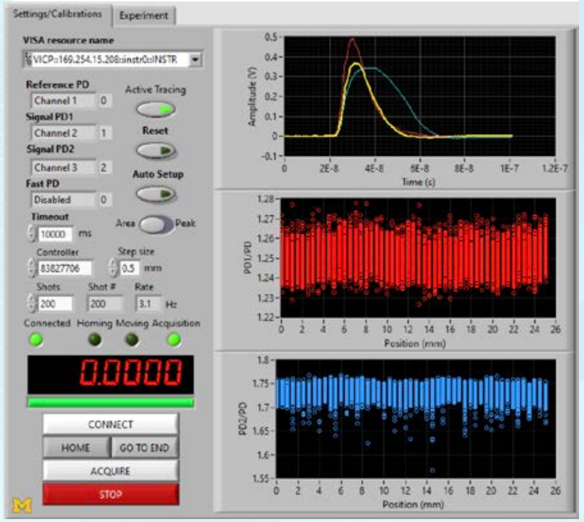
$$n_2 = \frac{12\pi^2 \chi^{(3)}}{n_0^2}$$



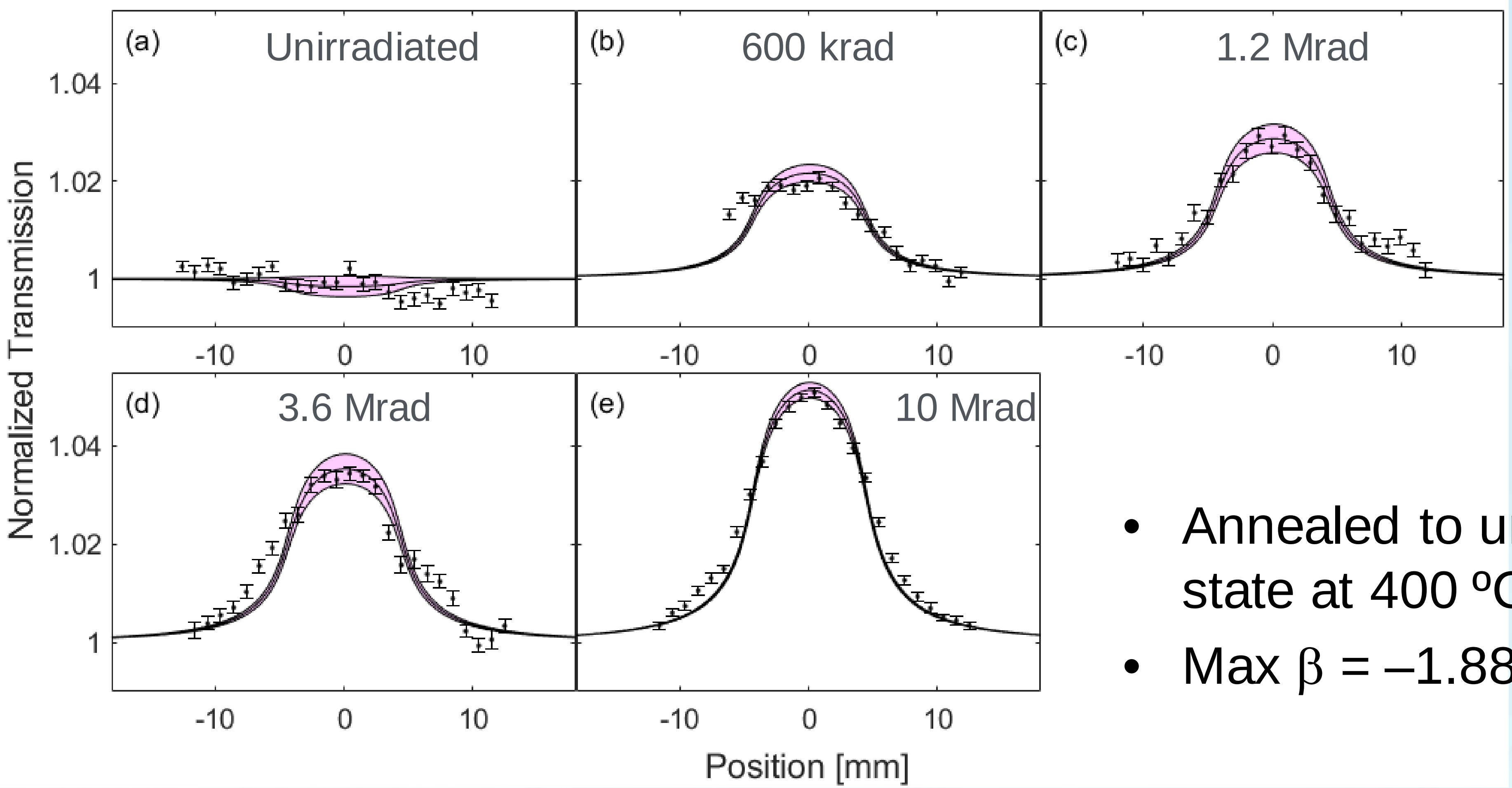
Measurement of Nonlinearities: Z-scan





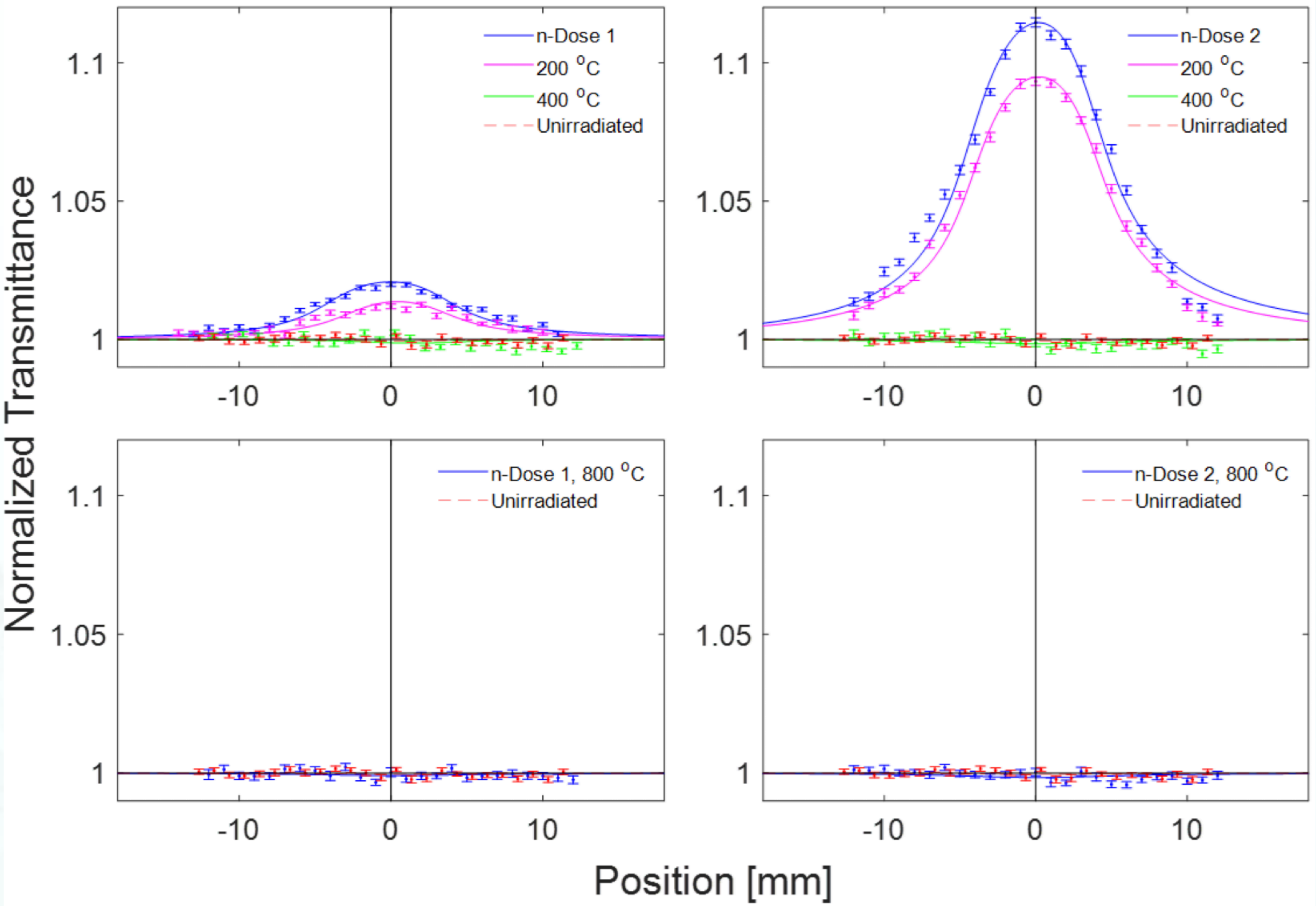


Observation of Negative Absorption in Quartz



- Annealed to unirradiated state at 400 ºC
- Max $\beta = -1.88 \times 10^{-13}$ m/W

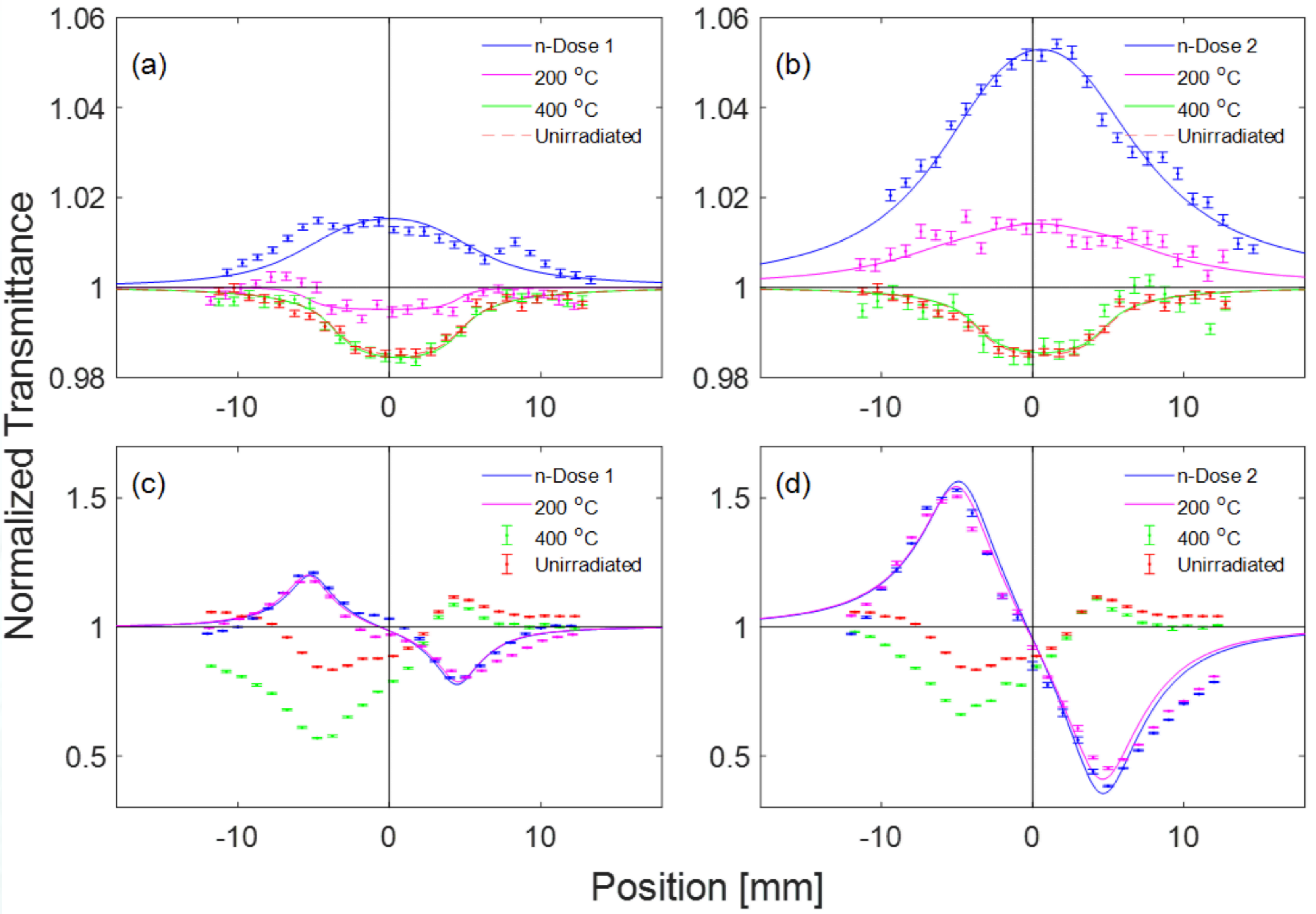
Neutron Irradiation Also Induces Negative Nonlinear Absorption



Sapphire

Annealing

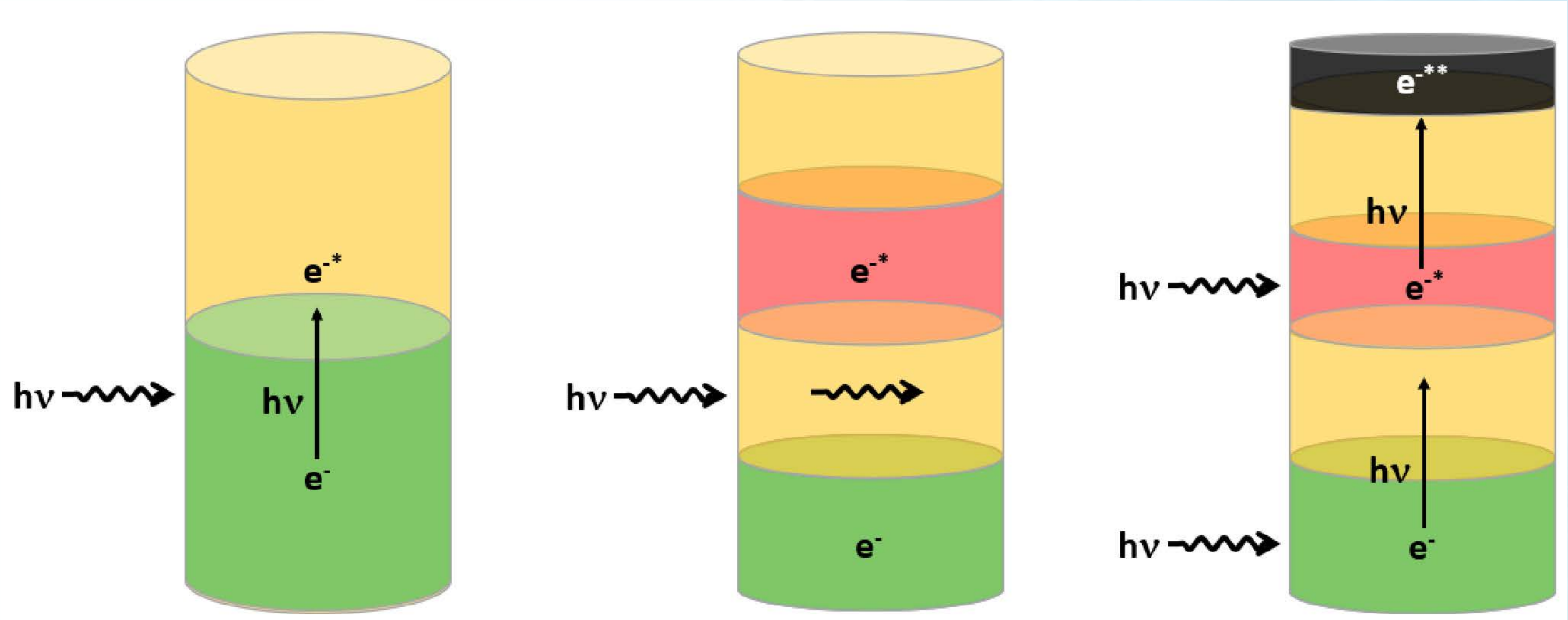
BK7G18 Neutron Irradiation: Nonlinear Absorption and Refraction



Nonlinear absorption

Nonlinear refraction

Negative Nonlinearity is Attributed to Saturated Absorption



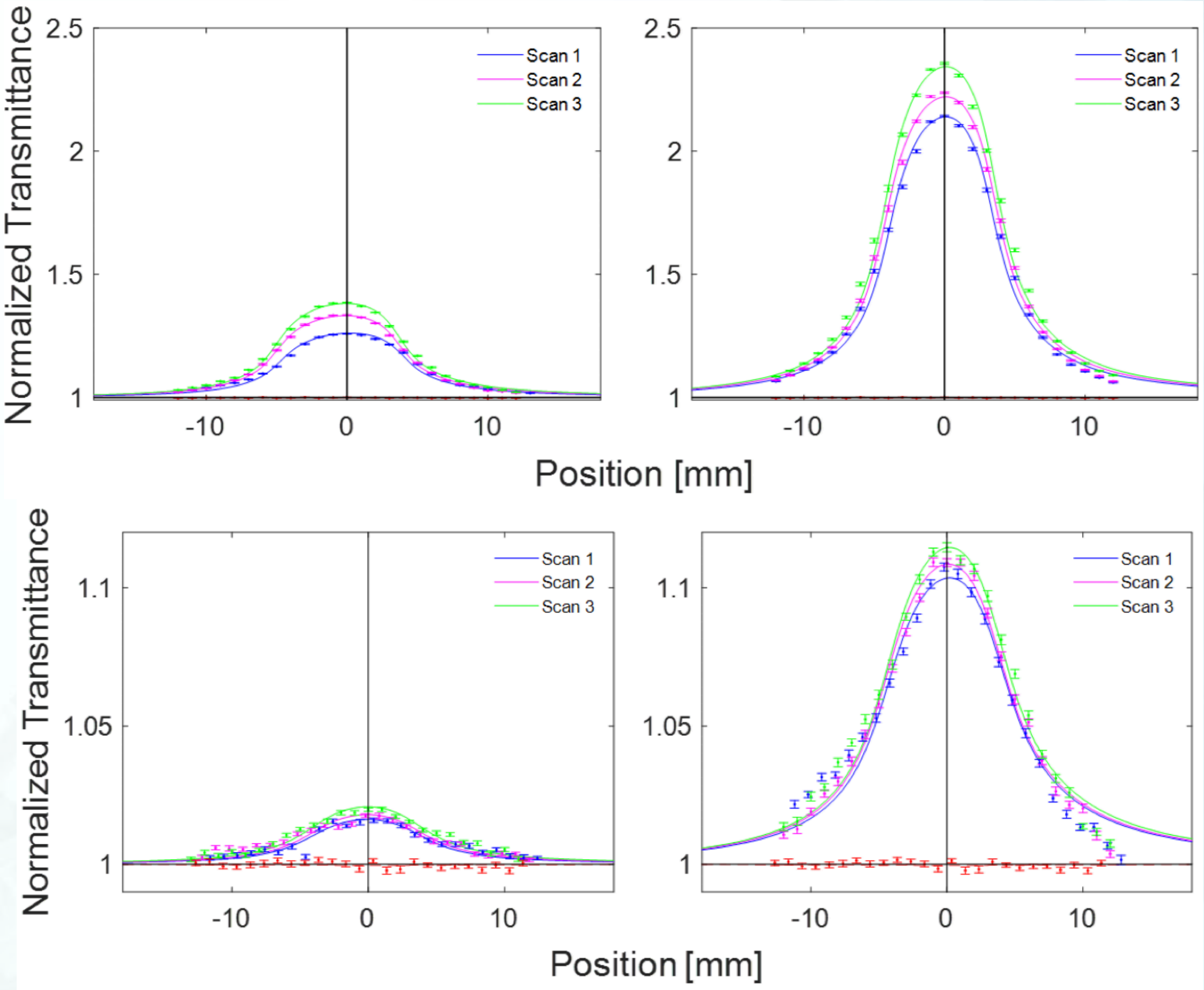
"Standard" linear absorption

Saturable absorption

Reverse saturable absorption

$$\alpha_{SA} = \frac{\alpha_0}{1 + I_0/I_S}$$

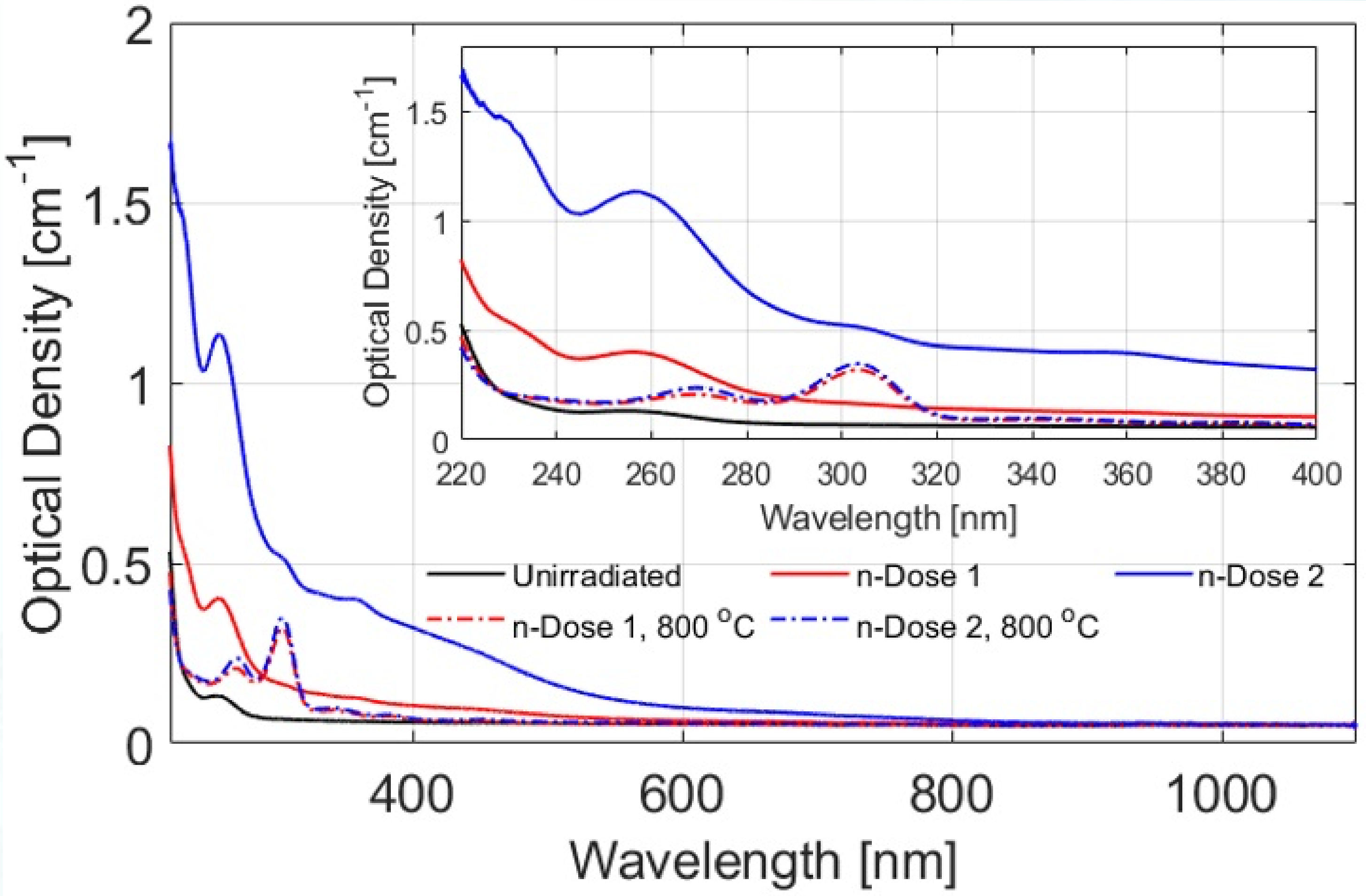
Photobleaching Measurements



Infrasil 302 low-OH fused silica: neutron irradiation (maximum observed effect)

Sapphire: neutron irradiation

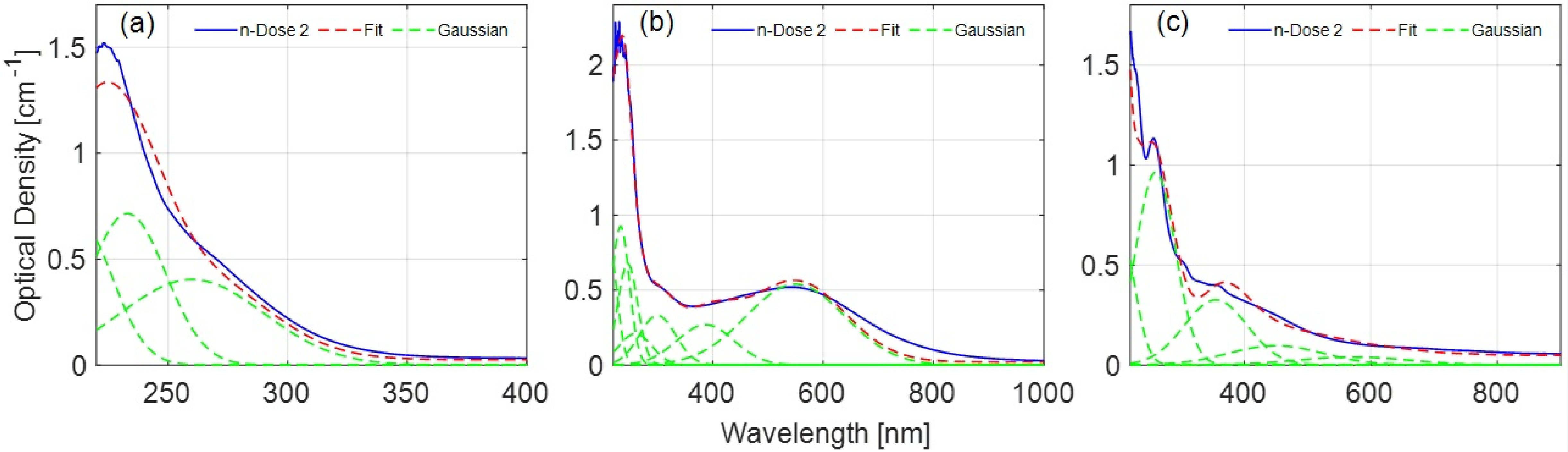
RIA in Sapphire with Concurrent Annealing



n-Dose 1: $3.4 \times 10^{16}\ n \cdot cm^{-2}$
n-Dose 2: $1.7 \times 10^{17}\ n \cdot cm^{-2}$

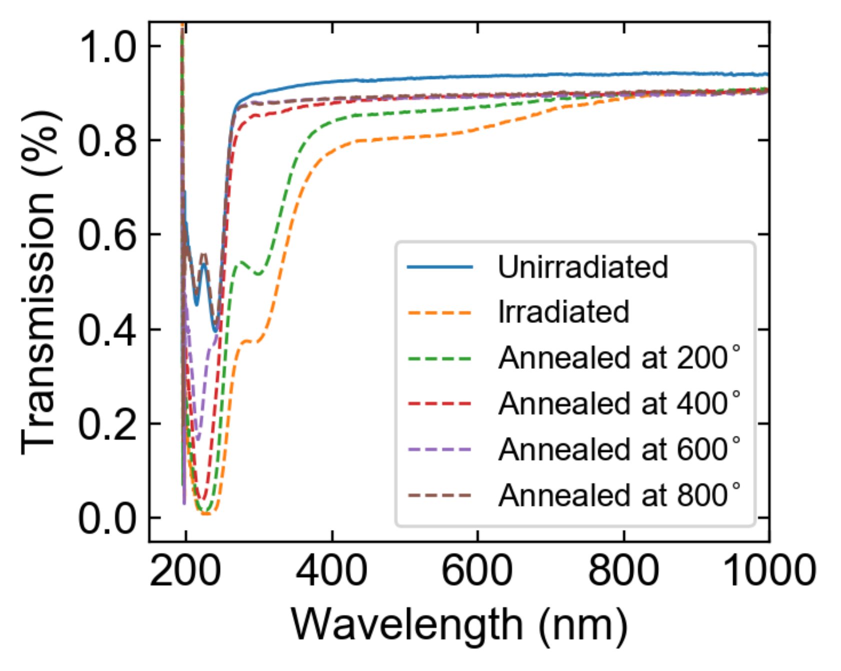
Features of Radiation-Induced Attenuation

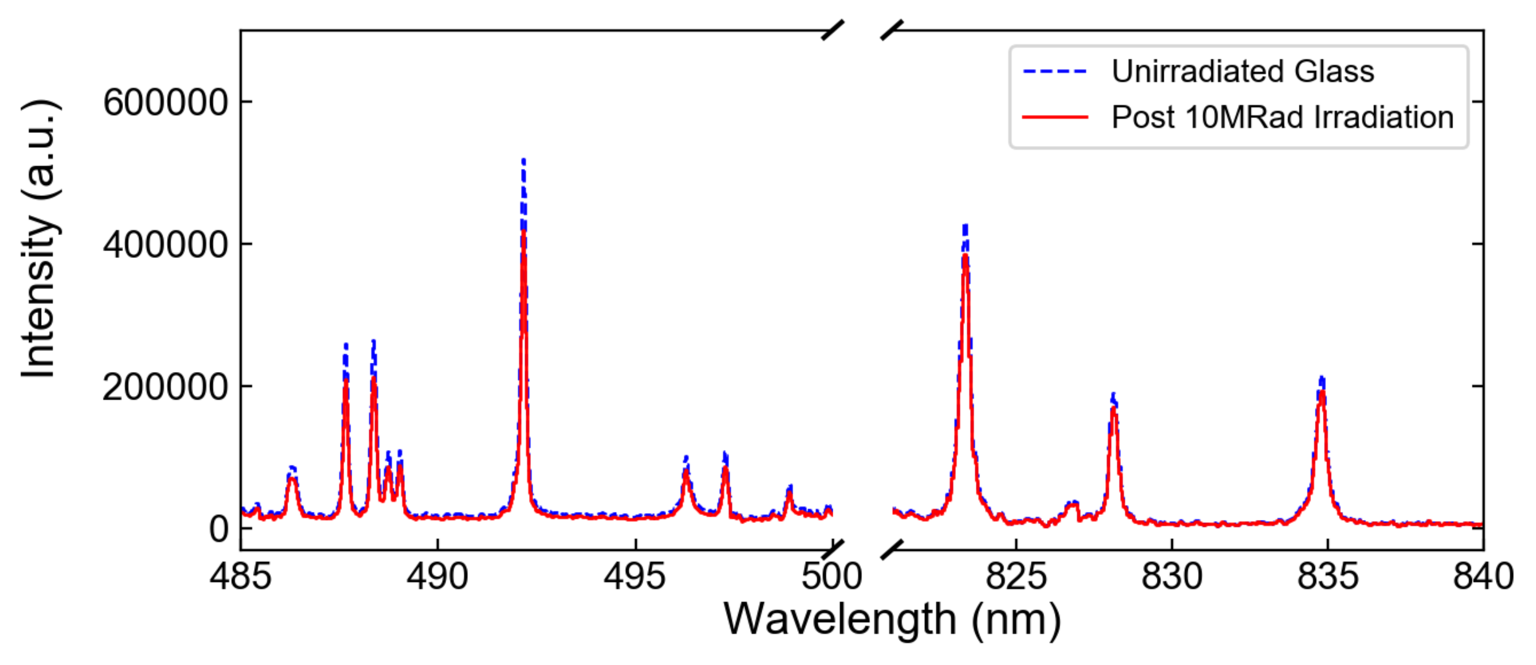
S2000 I302 Sapphire



Experimental data were fit with sums of Gaussians accounting for previously reported centroids and widths.

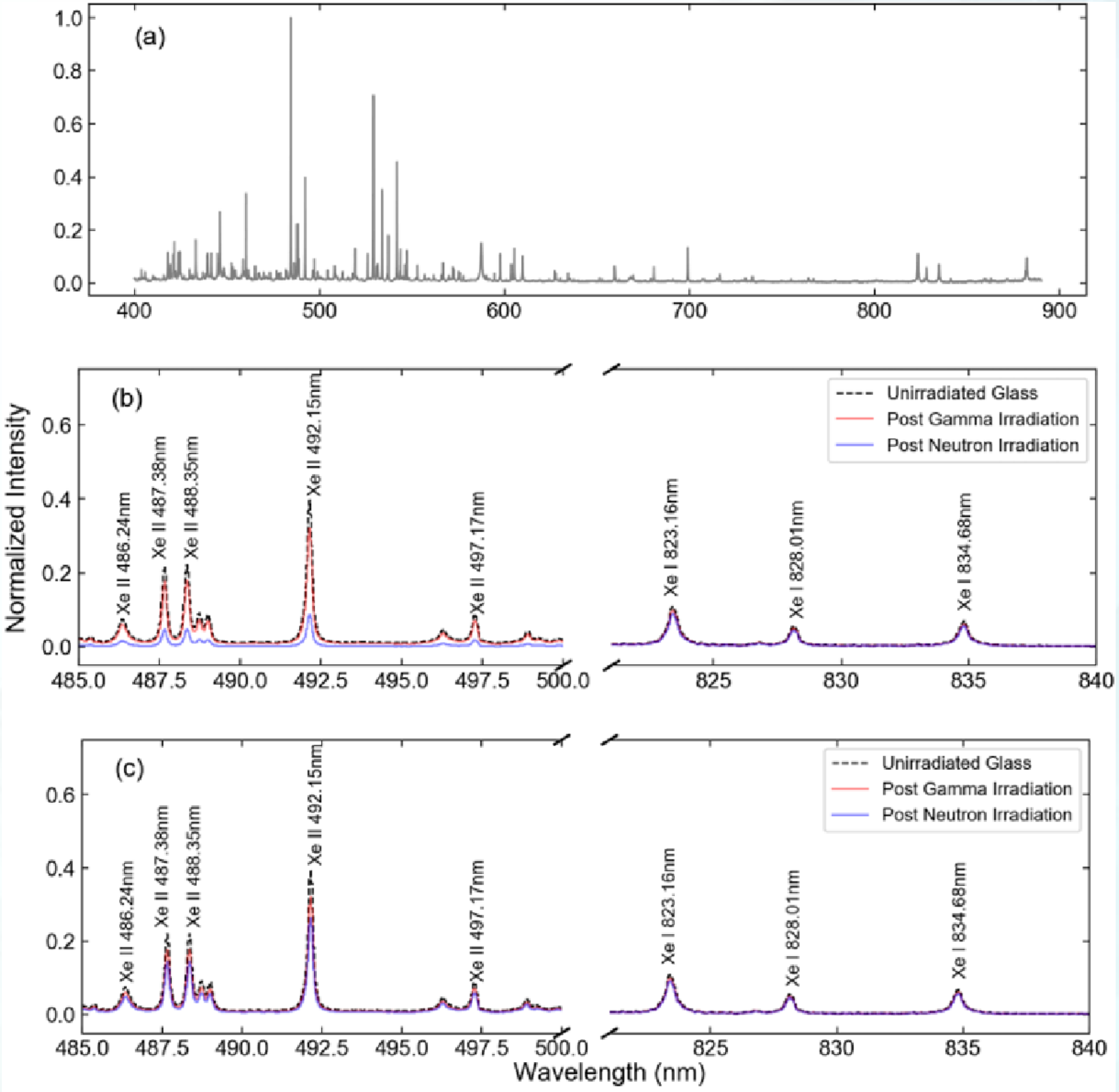
Effect of Irradiation on LIBS Instrumentation





Assumptions: LIBS with 1% Xe in He, 2 µs delay scaled by Infrasil-302, 10 MRad gamma irradiation data

Effect of Irradiation on LIBS Instrumentation



Xe + He LIBS spectrum

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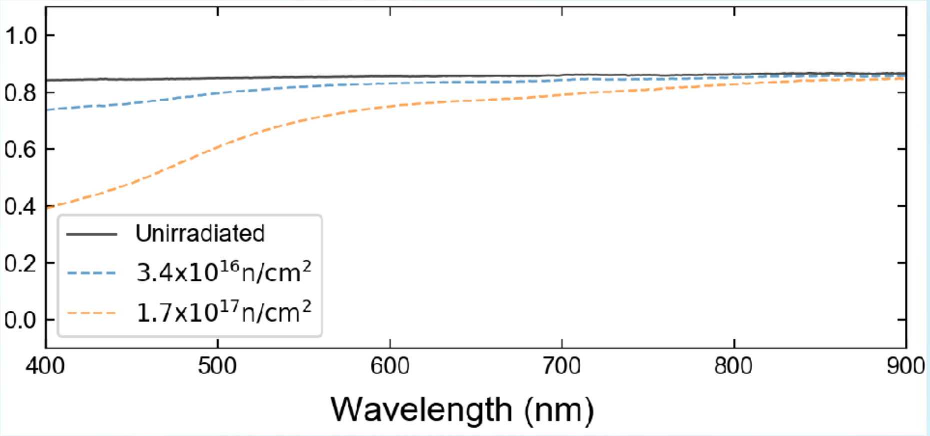
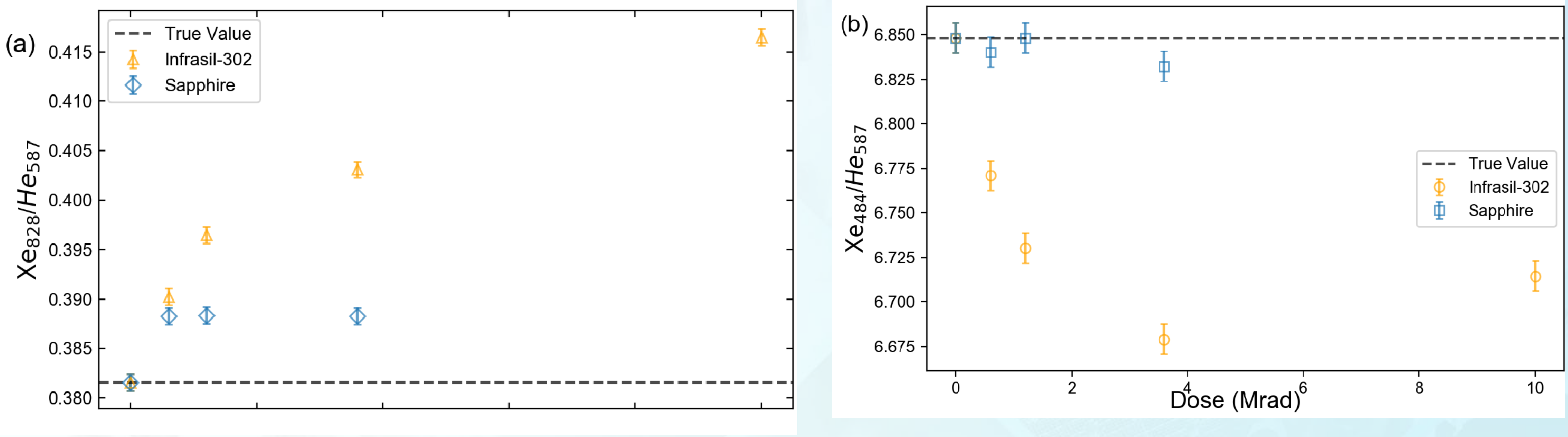


spectral line fitting

$$\frac{I(\lambda)}{I_0} = \frac{2}{\pi}\frac{\lambda_0}{w_l} I_\lambda \int_{-\infty}^{\infty} \frac{\exp\left[-\frac{2.772\lambda_0^2}{w_g^2}\left(\frac{v}{c}\right)^2\right] d\left(\frac{v}{c}\right)}{1+\frac{4}{w_l^2}\left[(\lambda-\lambda_0)-\lambda_0\left(\frac{v}{c}\right)\right]^2} + y_0$$

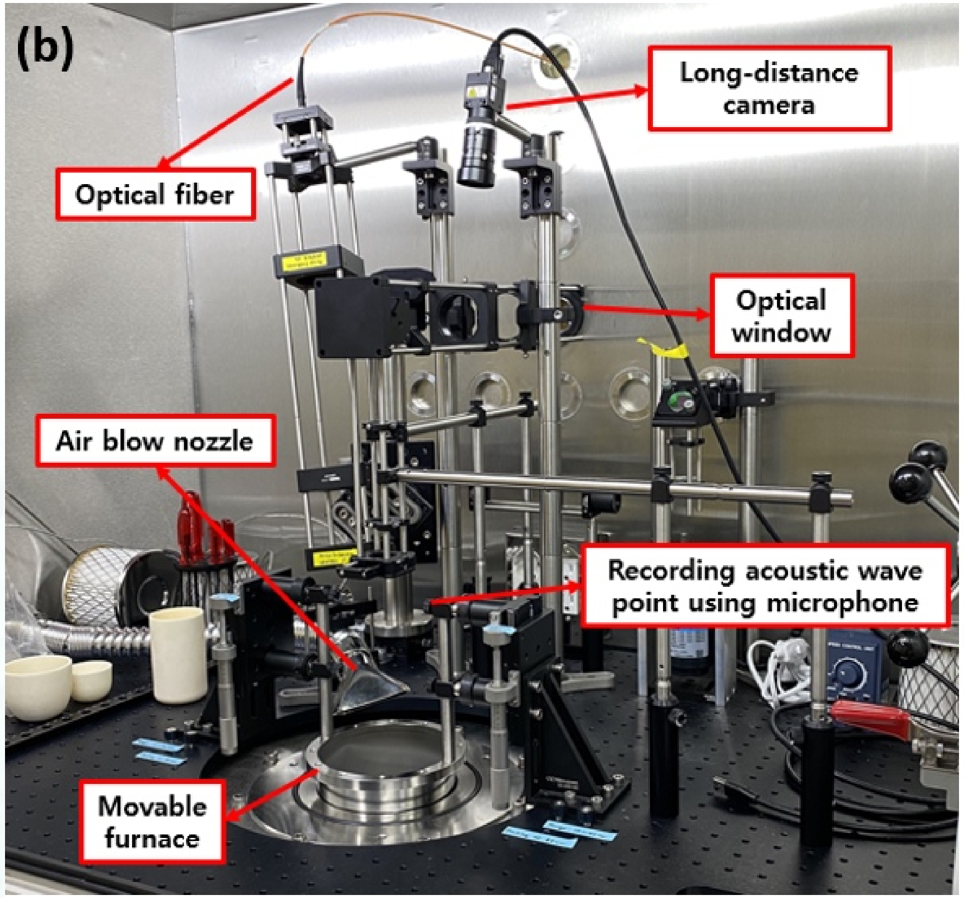
Effect of Irradiation on LIBS Instrumentation

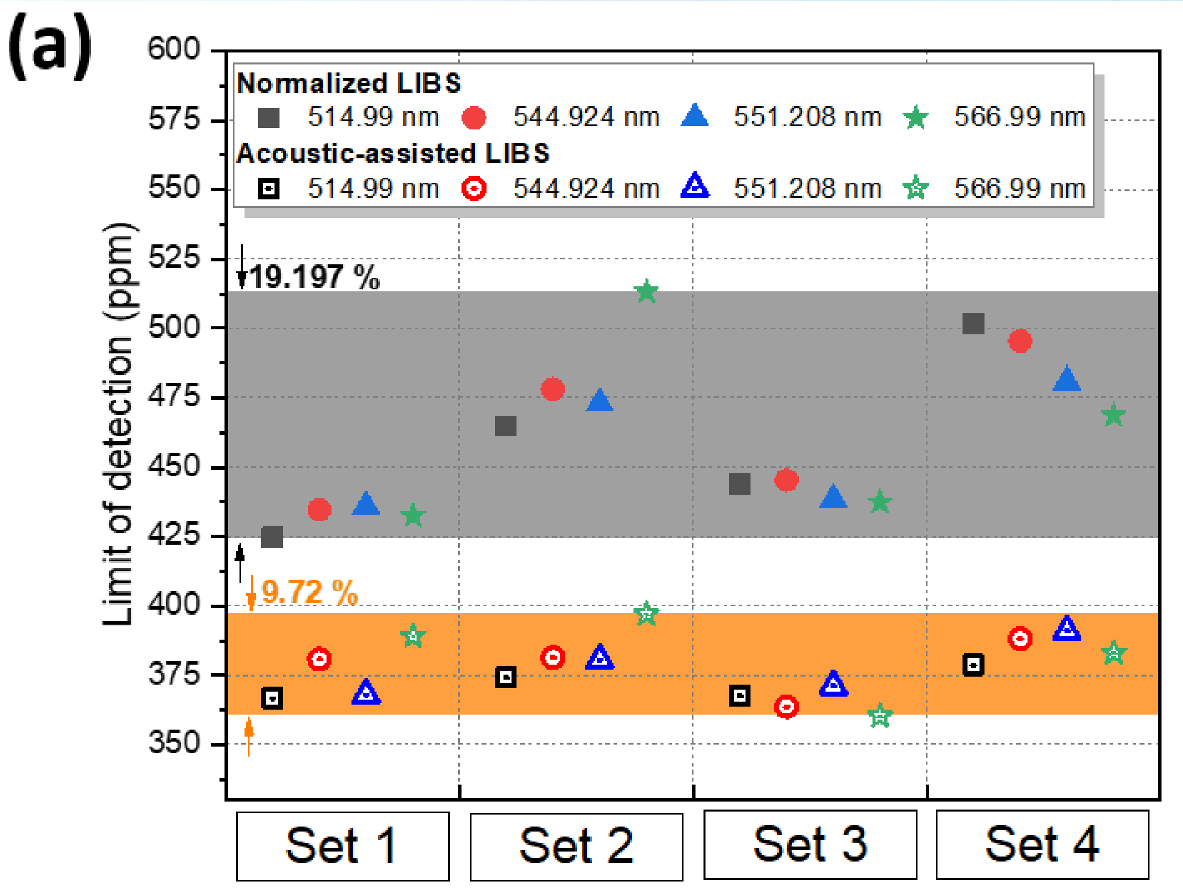


- Relative line intensities are used to determine relative component concentrations
- Irradiation can lead to systematic errors and calculation of plasma parameters used in normalization and correction for self-absorption.

INERI Collaboration (Seoul National University)

- Detection of corrosion products (Ni, Fe, Cr) in molten salts
- Cerium chloride (surrogate for plutonium) in LiCl-KCl eutectic
- Acoustic normalization





RTE Project Awarded (Collaboration with ORNL)

Radiation-induced Attenuation and Nonlinear Optical Properties of Fused Silica and Single-crystal Sapphire

- RIA and signal drift in a-SiO_2: reach an equilibrium?
- RIA in α-Al_2O_3 in samples irradiated to an even higher neutron fluence (~10^{22} n/cm^2) at high temperatures
- Radiation-induced changes of nonlinear optical properties of these materials

Rabbit	Dose (dpa)	Temperature (°C)	Material	ID	Dimensional	Archimedes	Optical transmission measurements	Dilatometry
OPT01	3.0	95	Al2O3	S02		X		X
OPT01	5.0	95	SiO2	H05				
OPT02	12.0	88	Al2O3	S06	X	X	X	X
OPT02	20.0	88	SiO2	H08	X		X	
OPT02	20.0	88	SiO2	H09	X		X	
OPT03	3.2	298	Al2O3	S10		X		X
OPT03	5.3	298	SiO2	H13				
OPT04	12.0	592	Al2O3	S14	X	X	X	X
OPT04	20.0	592	SiO2	H16	X		X	
OPT04	20.0	592	SiO2	H17	X		X	
OPT05	3.2	688	Al2O3	S18		X		X
OPT05	3.2	688	Al2O3	S19		X		
OPT05	3.2	688	Al2O3	S20		X		
OPT05	3.2	688	Al2O3	S21		X		
OPT05	5.3	688	SiO2	L18				X
OPT05	5.3	688	SiO2	H21				

OAK RIDGE National Laboratory

Journal Papers

- B. W. Morgan, M. P. Van Zile, C. M. Petrie, P. Sabharwall, M. Burger, and I. Jovanovic, “Radiation-Induced Negative Nonlinearities in Fused Silica, Sapphire, and Borosilicate Glass,” under preparation.
- L. J. Garrett, B. W. Morgan, M. Burger, Y. Lee, H. Kim, P. Sabharwall, S. Choi, and I. Jovanovic, “Impact of Glass Irradiation on Laser-Induced Breakdown Spectrocopy Diagnostics in the Visible in NIR Range,” in preparation.
- B. W. Morgan, M. P. Van Zile, C. M. Petrie, P. Sabharwall, M. Burger, and I. Jovanovic, “Optical Absorption of Fused Silica and Sapphire Exposed to Neutron and Gamma Radiation with Simultaneous Thermal Annealing,” *Journal of Nuclear Materials* 570, 153945 (2022).
- Y. Lee, S. Yoon, H. Kim, N. Kim, W. Yang, D. Kang, M. Burger, I. Jovanovic, and S. Choi, “In-situ measurement of Ce concentration in high-temperature molten salts using acoustic-assisted laser-induced breakdown spectroscopy with gas protective layer,” *Nuclear Engineering and Technology (2022)* https://doi.org/10.1016/j.net.2022.07.014.
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Gamma-radiation-induced negative nonlinear absorption in quartz glass,” *Optical Materials Express* 12, 1188-1197 (2022).
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Post-Irradiation Examination of Optical Components for Advanced Fission Reactor Instrumentation,” *Review of Scientific Instruments* 92, 105107 (2021).

Journal papers–under preparation Journal papers–published

Conference Presentations

- L. J. Garrett, B. Morgan, M. Burger, Y. Lee, H. Kim, S. Choi, and I. Jovanovic, “Impact of Glass Radiation Damage on Optical Spectroscopy,” ANS Winter Conference, Phoenix, AZ, November 13–17, 2022.
- B. Morgan, M. Burger, and I. Jovanovic, “Linear and Nonlinear Optical Properties of Fused Silica and Sapphire in Extreme Radiation and Thermal Environments,” IEEE Nuclear & Space Radiation Effects Conference, Provo, UT July 18–22, 2022.
- B. W. Morgan, M. Van Zile, P. Sabharwall, M. Burger, and I. Jovanovic, “Radiation-induced Negative Nonlinear Absorption in Glass and Sapphire,” Conference on Lasers and Electro-Optics, San Jose, CA, May 15-20, 2022.
- B. Morgan, M. Van Zile, P. Skrodzki, X. Xiao, P. Sabharwall, P. Marotta, M. Burger, and I. Jovanovic, “Post-Irradiation Examination of Irradiated Optical Components of In-Situ Spectroscopic Sensors for Advanced Fission Reactors,” ANS Winter Meeting, November 30–December 3, 2021.
- B. Morgan, P. Skrodzki, M. Burger, P. Sabharwall, P. Marotta, and I. Jovanovic, “Post-Irradiation Examination System Development for Irradiated Optical Components of In-Situ Spectroscopic Sensors,” ANS Winter Conference [online], November 15-19, 2020.

Summary of Accomplishments

- Mobile PIE system constructed and validated
- PIE system moved and operated at OSU NRL
- Constructed and operated thermal annealing furnaces
- Neutron/gamma irradiations completed; final analysis pending
- Established collaboration with ORNL, SNU, and UJM-SE
- New RTE project with ORNL
- INERI collaborative project with SNU
- 4 papers published; 2 papers under preparation; 5 conference presentations; 1 dissertation

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Summary of Accomplishments

Key research findings:

- Negative nonlinear absorption observed for the first time in bulk optical materials that include fused silica, sapphire, and BK7 crown glass
- Negative nonlinear refraction observed for the first time in BK7G18
- Additional information obtained on the ability of thermal annealing to repair radiation damage *in situ*

Future Work:

- True *in-situ* Z-scan can be performed using nuclear reactor beam ports
- Further investigation of the effect of photobleaching on nonlinear properties
- Measure the temporal duration of the saturable absorption
- Higher dose rates and temperatures – fusion conditions

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Thank You

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